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ScienceDirect

Scripta Materialia xxx (2008) xxx–xxx

Scripta MATERIALIA

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Tensile plastic deformation of a Zr-based bulk metallic glass composite in the supercooled liquid region

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Received 28 September 2008; revised 1 November 2008; accepted 4 November 2008

The tensile deformation behavior of $Zr_{55.9}Cu_{18.6}Ni_{10}Al_{7.5}Ta_8$ bulk metallic glass composite was investigated in the supercooled liquid region at various strain rates and temperatures. The deformation of the bulk metallic glass composite exhibited superplastic behavior, which is closely related to the strain rate and temperature. This excellent superplasticity, with a maximum elongation over 650%, indicates that the bulk metallic glass composite looks promising for thermoplastic forming applications.

Keywords: Bulk metallic glass composite; Tensile superplastic deformation; Deformation mechanism

Bulk metallic glasses (BMGs) have superior properties associated with their unique atomic structure, including high strength and hardness, large elastic limit, excellent corrosion and wear resistance [1–3]. In addition, some BMGs also exhibit superplasticity under external loading in the supercooled liquid region [4–12], which allows complicated near-net shape manufacturing of the glassy materials. However, monolithic BMGs are usually brittle in nature at room temperature, thus limiting their scope of application. Recently, it has been found that the formation of bulk metallic glass composite (BMGC) reinforced by a ductile crystalline phase is an effective approach to improve the plasticity of BMGs [13–15]. Using this method, a series of Zr-based BMGCs with enhanced global plasticity were fabricated, and some of them even exhibited a significant ductility in tension at room temperature [13].

In recent years, the deformation behavior of a few Zr-based BMGCs has been studied near the glass transition temperature or in the supercooled liquid region, under compression [16–18]. It has been found that BMGCs, just like monolithic BMGs, exhibited a large plasticity, but the deformation behavior is closely related to the temperature and to the strain rate. However, to the best of our knowledge, little work has been done on the tension of BMGCs in a supercooled liquid region. In this work, the tensile deformation of $Zr_{55.9}Cu_{18.6}Ni_{10}Al_{7.5}Ta_8$ BMG composite reinforced by Ta particles was systematically studied in the supercooled liquid region.

An alloy ingot with a nominal composition of $Zr_{55.9}Cu_{18.6}Ni_{10}Al_{7.5}Ta_8$ (at.%) was prepared by arc melting of pure metals (purity > 99.5%) in a Ti-gettered argon atmosphere. From the master alloy, a BMGC plate with a thickness of 1.5 mm was fabricated by copper-mold casting.

The structure and morphology of the as-cast alloy were examined by X-ray Diffraction (XRD; Philips 1860) and scanning electron microscopy (SEM, Quanta 200), respectively. Both XRD pattern and SEM morphology revealed that the BMGC reinforced by granule Ta crystallites had formed (see Fig. 1). The thermal response of the alloy was investigated by differential scanning calorimetry (DSC, Perkin-Elmer DSC-7) at a heating rate of 20 K min^{-1}, which revealed a glass transition temperature of 669 K with a supercooled liquid region of 94 K.

The uniaxial tensile test was performed at various temperatures in the supercooled liquid region (between 683 and 713 K) and different strain rates (between 1×10^{-3} and $1 \times 10^{-2}\ s^{-1}$) using an Instron machine

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doi:10.1016/j.scriptamat.2008.11.006

Please cite this article in press as: S.F. Guo et al., Scripta Mater. (2008), doi:10.1016/j.scriptamat.2008.11.006

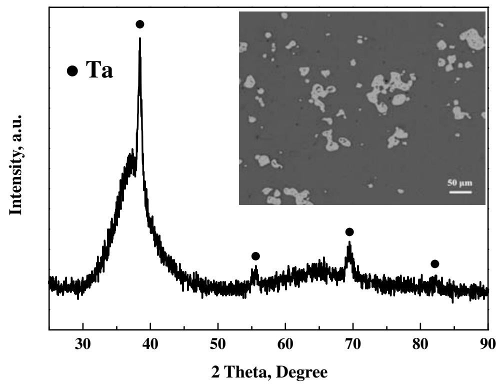

Figure 1. XRD pattern and backscattered scanning electron micrograph of as-cast $Zr_{55.9}Cu_{18.6}Ni_{10}Al_{7.5}Ta_8$ BMGC plate.

equipped with an air furnace. Samples for the tensile test with a gauge length of 9.5 mm and gauge width of 1.5 mm were fabricated by wire-cutting from the as-cast plate. In order to shorten the time for heating the sample, the load train was preheated to the test temperature; a sample was then rapidly placed into the load train and held for another 8–10 min to stabilize its temperature. The fluctuation of the temperature in the furnace during testing was about ±3 K.

Figure 2a shows the strain-rate dependence of the true stress–true strain (σ–ε) curves of the BMGC at a certain temperature of 693 K (for simplification, only three curves are presented in the figure). All σ–ε curves are characterized by a stress overshoot in the initial stage of deformation followed by a steady-state flow with a stress plateau in the extended strain region. The peak of the stress-overshoot increased as the strain rate increased, while the steady-state stress seems independent of the strain rate with an average value of about 25 MPa. Interestingly, the σ–ε curve at the strain rate of 1×10^{-3} (the lowest strain rate used in the study) was characterized by a number of serrations and shows evidence of strain-hardening during the later stage of the deformation. Similar results were also reported previously in monolithic BMGs [9]. The strain hardening is most likely attributable to the concurrent crystallization in the extended process of deformation, which was confirmed by the subsequent XRD examination. The overall appearance of the BMGC samples deformed at different strain rates is given in the insert of the figure. As compared with the undeformed sample, all the deformed BMGCs exhibit a superplastic behavior with a maximum elongation of 600% at $5 \times 10^{-3}\ s^{-1}$.

Figure 2b shows the temperature dependence of the σ–ε curves of the BMGC at a fixed strain rate of $5 \times 10^{-3}\ s^{-1}$. Similar to the strain rate effect, the deformation behavior is also greatly dependent on the temperature. The stress overshoot appeared at the low temperature (e.g. 683 K), but the peak of stress overshoot gradually decreased with the increase of the temperature and finally disappeared when the temperature was higher than 698 K. The BMGC also shows a large elongation in this case with a maximum value of 655% at 698 K, as shown in the insert of Figure 2b. Although the deformation behavior of the BMGC shows a close dependence on the strain rate and temperature, the tensile elongation in all cases exceeds 200%. This demonstrates that the BMG composite, just like some monolithic BMG, has a very good deformability in the supercooled liquid region.

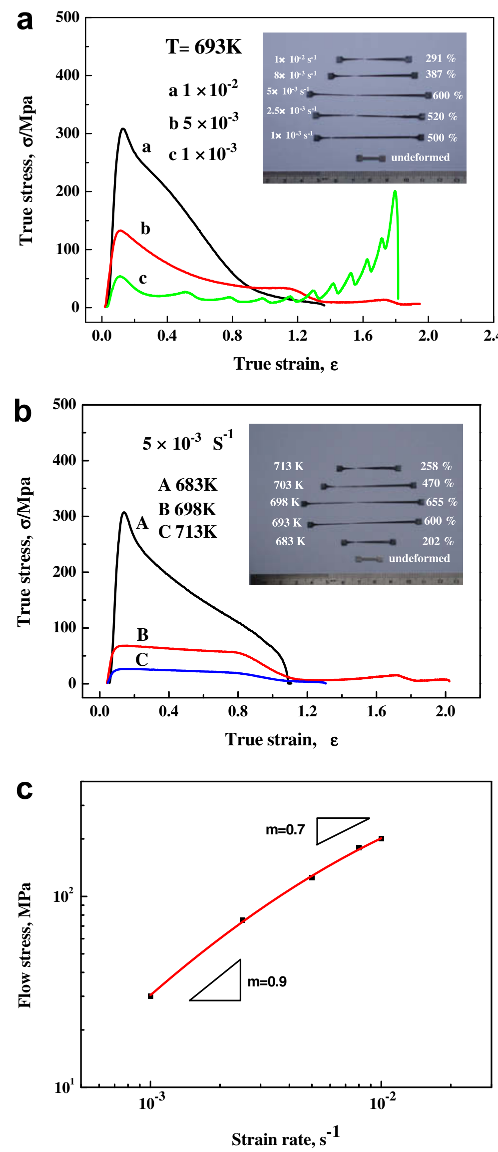

Figure 2. (a) The strain-rate dependence of the true tress–true strain curves at 693 K for the BMGC; (b) the temperature dependence of the true tress–true strain curves at a fixed strain rate of $5 \times 10^{-3}\ s^{-1}$; (c) the logarithmic relationship between the flow stress and the strain rate at 693 K for the BMGC.

Figure 2c shows the logarithmic relationship between the nominal flow stress and the strain rate at the temperature of 693 K. Here the nominal flow stress was defined using the method proposed by Kawamura [19]. It can be seen that the flow stress increased monotonously as the strain rate continued to increase. The slope of a straight line actually reflects the value of strain rate sensitivity, m, according to the definition of $m = \partial \log \sigma_{flow} / \partial \log \dot{\varepsilon}$. The m value is close to unity at a strain rate lower than $5 \times 10^{-3}\ s^{-1}$, implying that the plastic deformation of the BMGC behaves nearly in a Newtonian flow in a low strain rate regime. However, m decreases steadily with an increasing strain rate, indicating a transition from a Newtonian-to-non-Newtonian state in the high strain rate regime. The similar transition from Newtonian-to-non-Newtonian behavior as the strain rate increases has been observed previously in the compression of the BMGC [17]. It is noted that the m value is always higher than 0.7 within the strain rate range used in the study. It is believed that the high m value is responsible for the large elongation of the BMGC.

Figure 3 shows a SEM micrograph of one deformed sample with an elongation of 600% (deformed at $5 \times 10^{-3}\ s^{-1}$ and 693 K), which shows clearly an orientated distribution of Ta particles along the tensile direction after deformation. The result implies that the deformation of BMGC in the supercooled liquid region is dominated by the viscous flow of the BMG matrix. Actually, this is quite reasonable, because Ta particles only possess a minor portion (~10% in volume) in the BMG composite and they are much harder than the viscous amorphous matrix in the supercooled liquid region. Therefore, the Ta particles may not carry much load during the deformation and are not able to affect significantly the deformation behavior of the BMG composite.

The above results show that the deformation behavior of the Zr-based BMGC in the supercooled liquid region strongly depends on the strain rate and temperature. The alloy exhibited Newtonian-like behavior in the lower strain rate regime, but became non-Newtonian in the high strain rate regime. The transition state theory, which can predict the correlation between strain rate and flow stress of a material, is extensively used to explain the transition from a Newtonian flow to a non-Newtonian flow in various BMGs [20–22]. The simplified description of the constitutive law can be written as [17,23,24]:

$$\dot{\varepsilon} = \dot{\varepsilon}_0 \sinh\left(\frac{\sigma V_{act}}{2\sqrt{3}kT}\right) \quad (1)$$

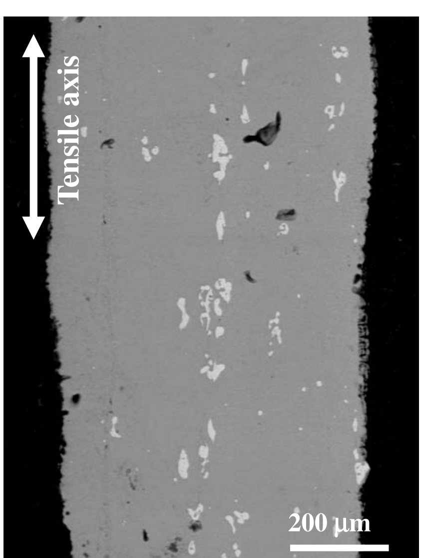

Figure 3. Scanning electron micrograph of the BMGC specimen deformed at $5 \times 10^{-3}\ s^{-1}$ and 693 K, which shows a structural orientation along the tensile direction.

where T is temperature at which the deformation is carried out, k is Boltzmann's constant, the activation volume V_{act}, as well as the reference strain rate $\dot{\varepsilon}$ are used as fitting parameters for adapting the model curves to the experiment data. As shown by the broken line in Figure 4, the date can be fitted quite well with the constitutive law using the pre-exponential factor $\dot{\varepsilon}_0 = 3.92 \times 10^{-3}\ s^{-1}$ and the activation volume $V_{act} = 274\ Å^3$. This value of activation volume is found to be much higher than those reported previously for other monolithic Zr-based BMGs [24,25]. The large activation volume was also found in our previous work dealing with compression [17]. It is speculated that the large activation volume in BMGC in the present case probably results from the high defect concentration around the boundaries between Ta particles and the amorphous matrix because of a large difference in thermal mechanical mismatch (such as elastic modulus and thermal expansion coefficient) and chemical incompatibility between the two phases. The higher density of defects can increase the atomic diffusion [26], which in turn enhances plastic flow of materials during the deformation process. It is worthy of mentioning that the steady flow stress in tension is only around 25 MPa, which is very low and should be beneficial for superplastic forming. This result supports the viewpoint that the existence of micro-scale particles does not decrease the superplasticity of the BMG in the supercooled liquid region, at least, in the case where the micro-scale crystalline phase was kept to an appropriate amount.

In summary, the superplastic deformation of $Zr_{55.9}Cu_{18.6}Ni_{10}Al_{7.5}Ta_8$ BMG composite in a supercooled liquid region under tension was investigated for the first time. The BMG composite exhibits an excellent superplasticity over a wide range of strain rates and temperatures with a maximum elongation over 650%. The deformation behaves in the manner of a Newtonian flow in the low strain rate regime, but becomes non-Newtonian in the high strain rate regime. The deformation of the BMG composite in the supercooled liquid region can be well fitted by the transition state theory. The excellent superplasticity of the BMG composite demonstrates that this type of material is also promising in the application of thermoplastic forming.

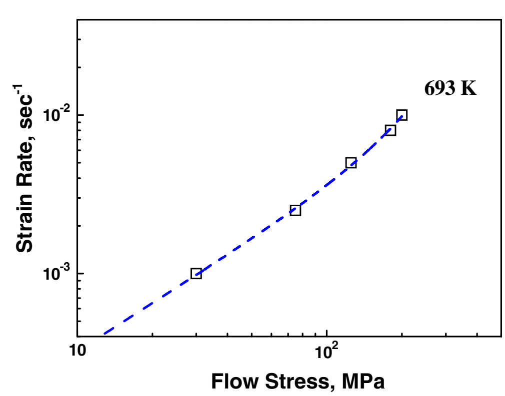

Figure 4. Fit of Eq. (1) (broken line) with experimental data (square) for $Zr_{55.9}Cu_{18.6}Ni_{10}Al_{7.5}Ta_8$ BMGC.

Please cite this article in press as: S.F. Guo et al., Scripta Mater. (2008), doi:10.1016/j.scriptamat.2008.11.006

This work was financially supported the by the Research Grant Council of The Hong Kong Special Administration Region (Project No. PolyU5254/04E), and partially supported by the Natural Science Foundation of China under Grant No. 50635020. The authors are grateful to the Analytical and Testing Center, HUST, for technical assistance.

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